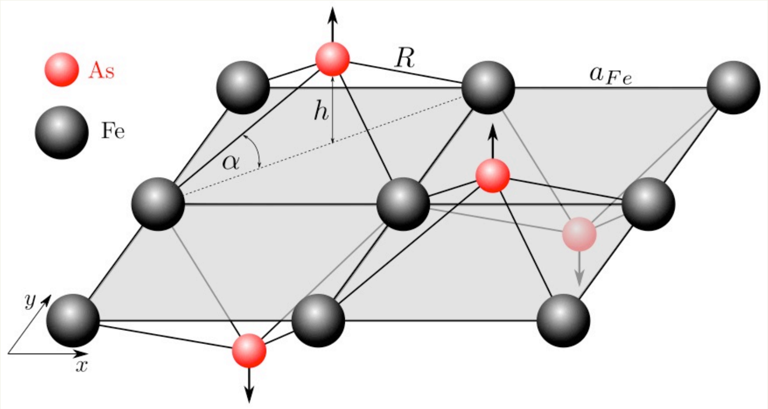
Coupling of the A1g As-phonon to magnetism in iron pnictides

Belén Valenzuela

Instituto de Ciencias Materiales de Madrid

(ICMM-CSIC)



BNL, New York 2013

In collaboration with:

Noel A. García-Martinez
(ICMM-CSIC)

Sergio Ciuchi
(Universitá dell'Aquila)

Emmanuele Cappelluti
(CNR Roma)

Maria J. Calderón
(ICMM-CSIC)

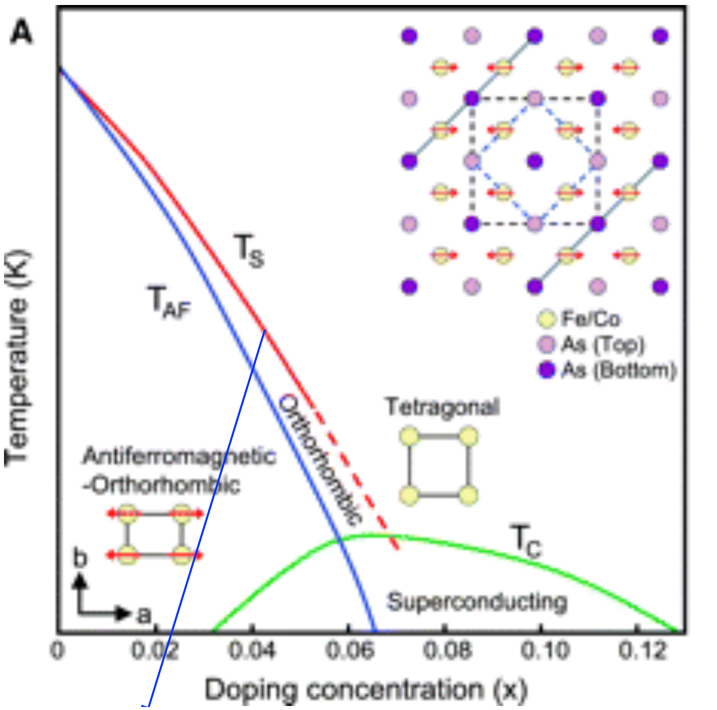
Elena Bascones
(ICMM-CSIC)

Index

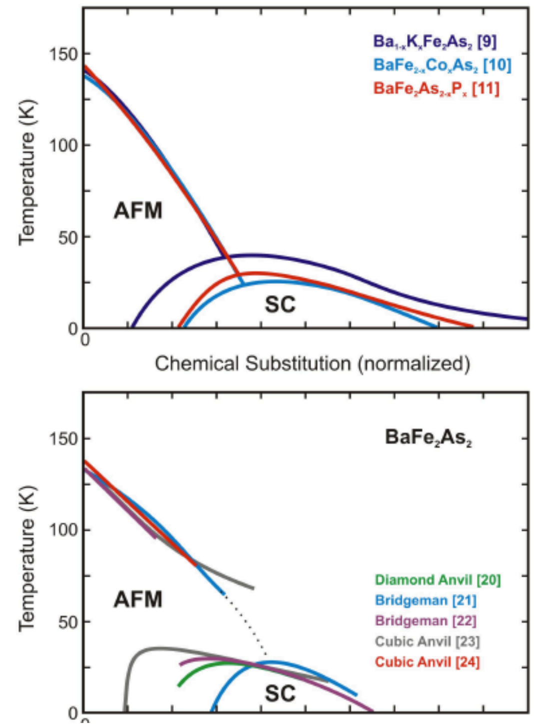
- Introduction: Interplay of charge, spin and lattice dynamics in iron superconductors
  - A1g As-phonon Raman response
- Our work:
  - Electron-phonon Hamiltonian plus interactions.
  - Phonon Raman response using charge-phonon theory.
  - Results: Raman intensities of the A1g As-phonon in the A1g, B1g and B2g symmetries.Hardening/ softening.

Charge-spin-lattice entanglement in iron superconductors

Davis, et al. Science'10



Green&Paglione, Nat.Phys.'10



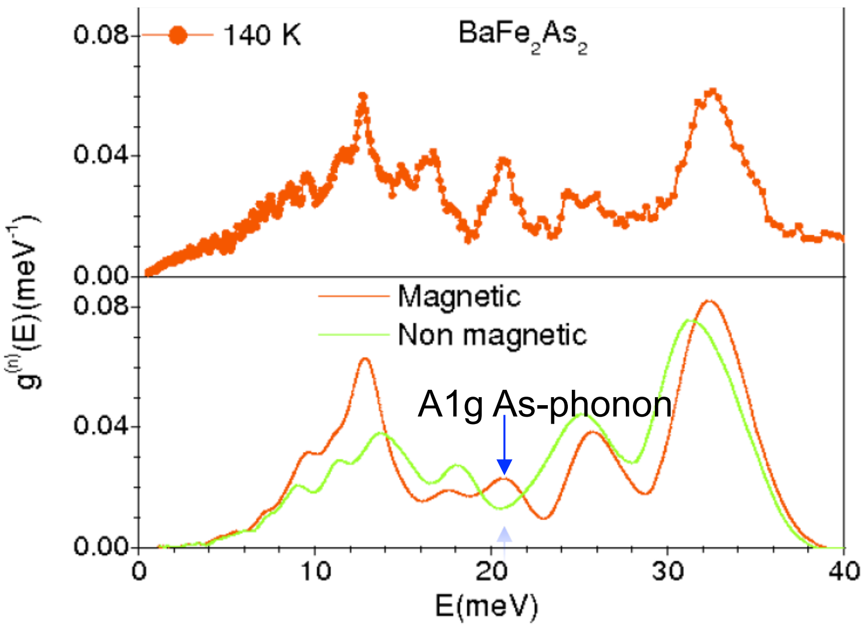
The orthorhombic transition follows the magnetic transition with **(π,0)** ordering

Both, chemical doping and pressure suppress the SDW and induce SC.

BNL, New York 2013

Charge-spin-*phonon* entanglement in iron superconductors

- Experimentally several phononic signatures show unconventional behavior in the magnetic state: Choi et al. PRB'08, Le Tacon et al. PRB'08, PRB'09, Chauviere et al. PRB'09, PRB'11, Akrap et al. PRB'09, Zhang et al. JACS'10, Schafgans et al. PRB'11, Nakajima et al. PNAS'11, Kim et al Nat. Mat.'12, Liu et al. PRL'13…
- Theoretically (Ab-initio calculations):

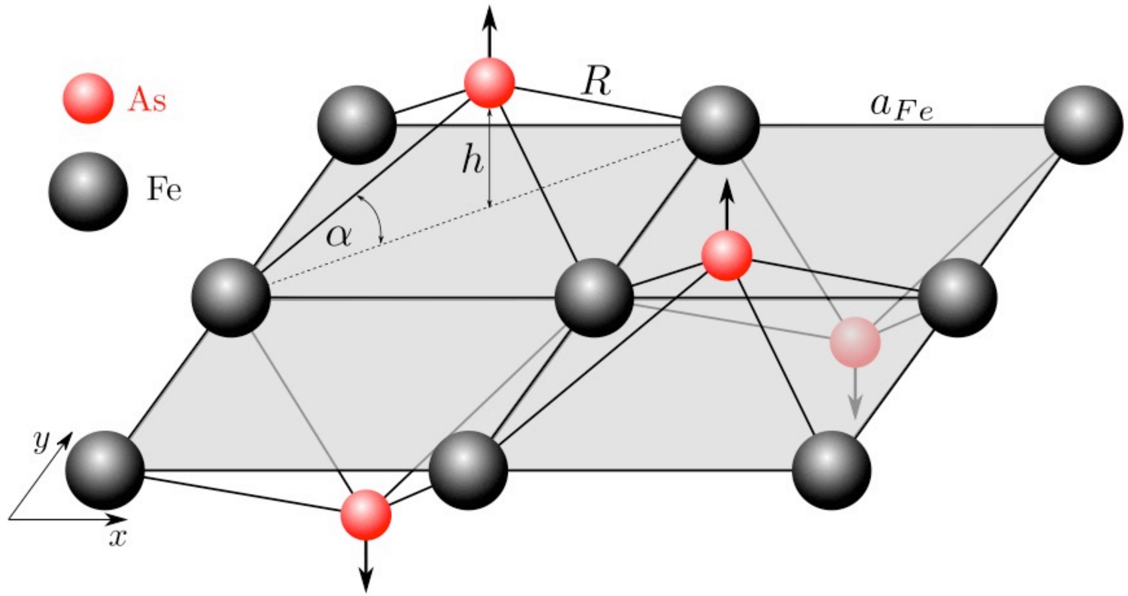


Zbiri et al., Phys. Cond. Matt'10

- iron-magnetism is present also above T_N? what about the estimations of the electron-phonon coupling?
- The electron-phonon coupling has been shown to be enhanced by magnetism. (Yndurain et al PRB'09, Boeri et al. PRB'10)
- Role for the mechanism of superconductivity?

BNL, New York 2013

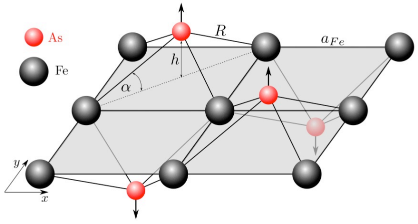
As height

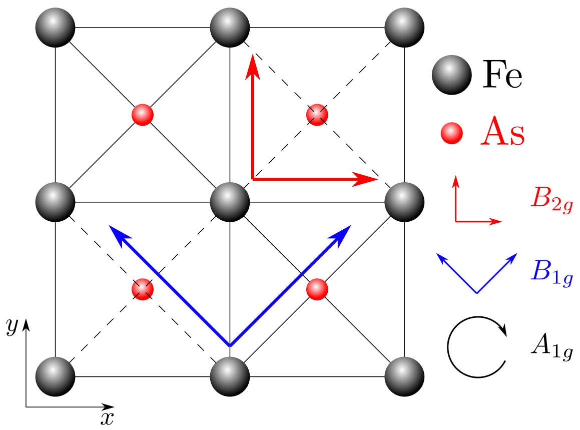


It has been claimed that the height of the As atom affects:

- the band structure at the Fermi level
- the magnetic moment, magnetic ordering
- the superconducting critical temperature, sc gap

Raman A1g As-phonon

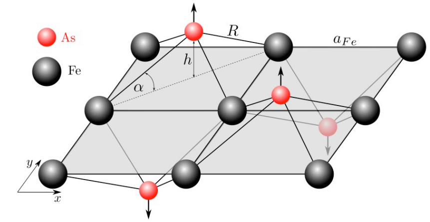


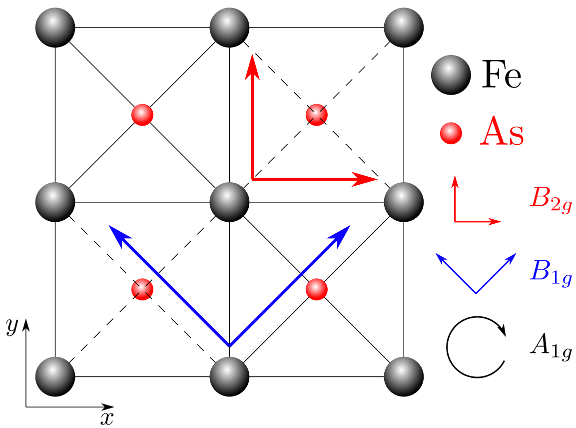


A1g=LL
B1g=x'y'
B2g=xy

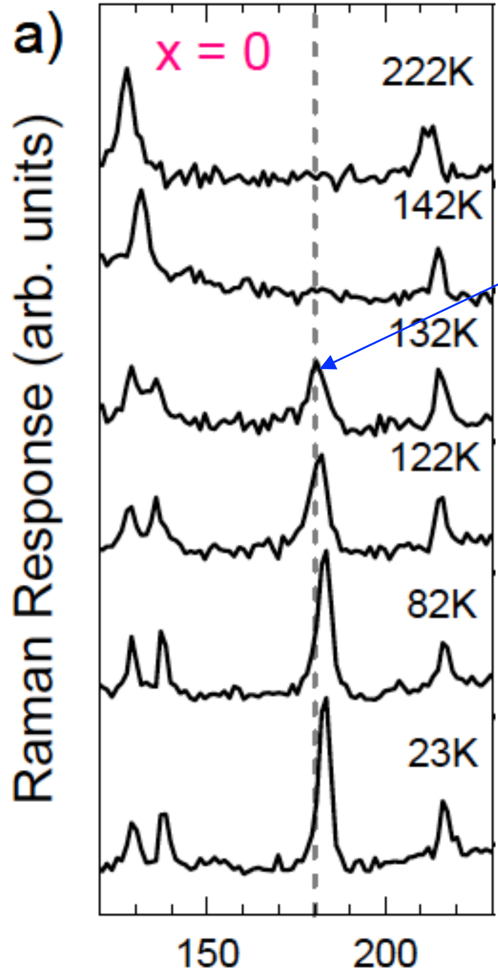
BNL, New York 2013

Raman A1g As-phonon





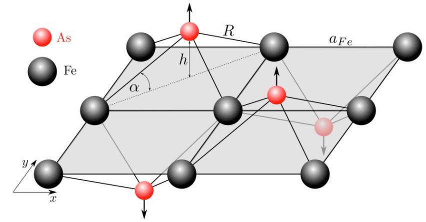
A1g=LL
B1g=x’y’
B2g=xy

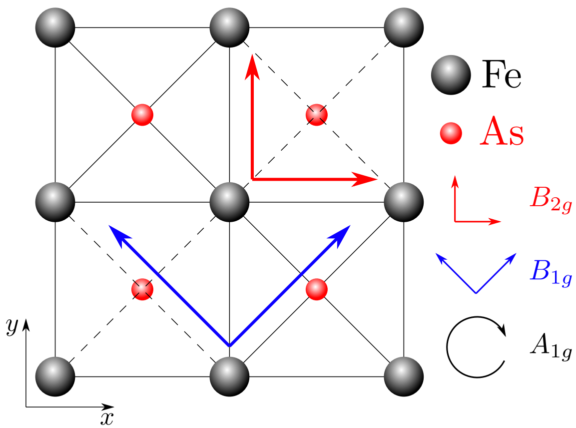


Chauviere et al PRB’11
($BaFe_2As_2$)

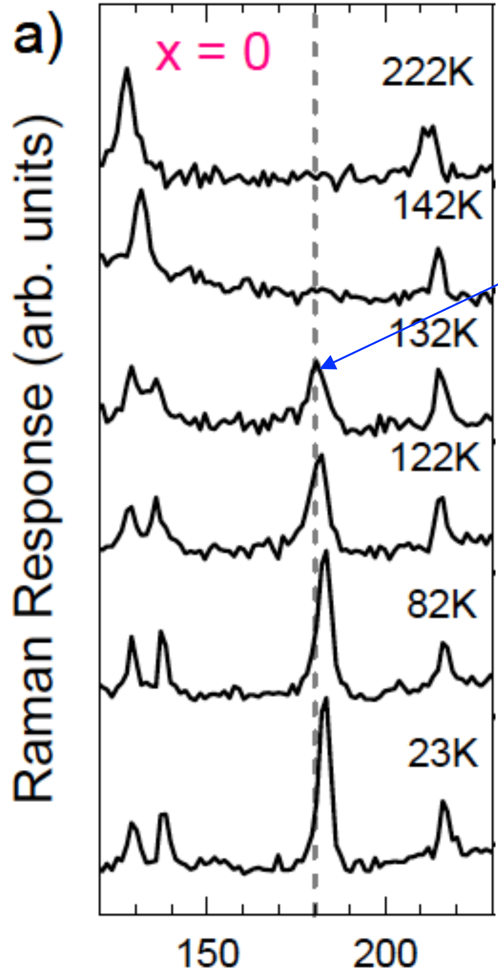
Below the Magneto-structural transition the **A1g As-phonon Intensity increases a lot for B1g (x’y’)**

Raman A1g As-phonon



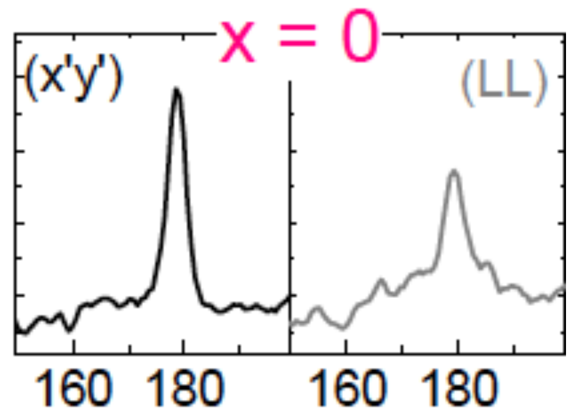


A1g=LL
B1g=x'y'
B2g=xy



Chauviere et al PRB'11 ($BaFe_2As_2$)

Below the Magneto-structural transition the **A1g As-phonon Intensity increases a lot for B1g (x'y')**

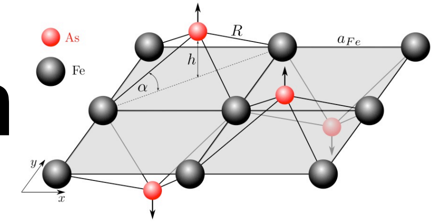


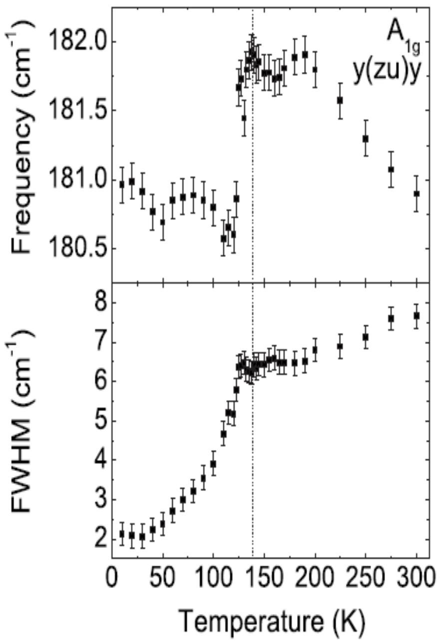
A1g As-phonon intensity in B1g is 1.5 bigger than in A1g in the magnetic state of $BaFe_2As_2$

The structural transition cannot explain these features, Why then?

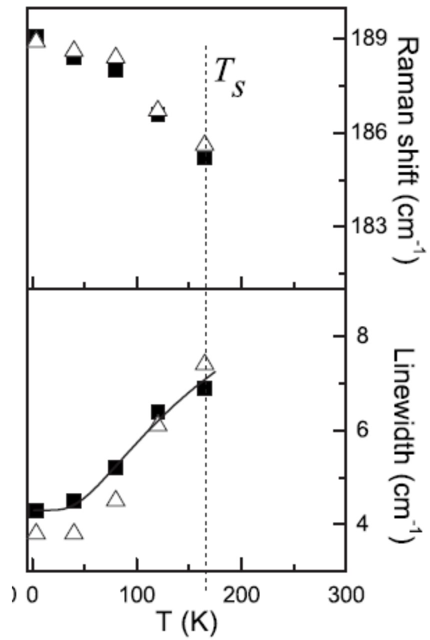
BNL, New York 2013

Raman A1g As-phonon





Rahlenbeck et al PRB'09 ($BaFe_2As_2$): Below the Magneto-structural transition **softening** and narrower scattering rate



Choi et al PRB'08 ($CaFe_2As_2$): Below the Magneto-structural transition **hardening** and narrower scattering rate

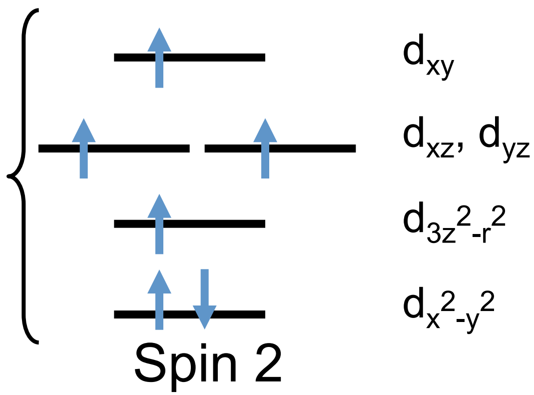
BNL, New York 2013

Our work:
Coupling of the A1g arsenide phonon to magnetism in iron pnictides

N. García-Martinez, B.V, M.J. Calderón, S. Chiuci, E. Cappelluti, E. Bascones,
arXiv:1307.7065

Our model: Microscopic Hamiltonian

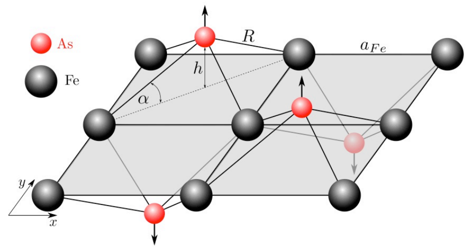
6 electrons in 5 d orbitals in a tetrahedral environment with crystal field 100-200meV:



Introducing on-site interactions:
U -> intraorbital repulsion
U' -> interorbital repulsion
J -> Hund's coupling
U'=U-2J

$$H = H_{TB} + H_{\mathrm{ph}} + H_{\mathrm{int}}$$

Tight-binding



$$H_0 = \sum_{\mathbf{k},\mu,\nu,\sigma,r} F^r_{\mu\nu}(\mathbf{k}) t^r_{\mu\nu} c^\dagger_{\mathbf{k}\mu\sigma} c_{\mathbf{k}\nu\sigma} + \sum_{i,\mu,\sigma} \epsilon_\mu c^\dagger_{i\mu\sigma} c_{i\mu\sigma}$$

✓ Focus on Fe-pnictogen planes, square lattice, Fe unit cell

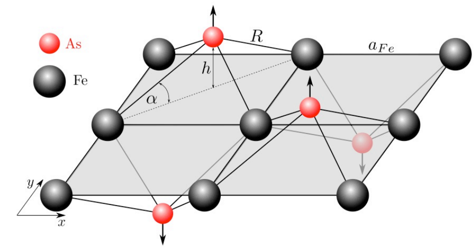
✓ Five Fe d-orbitals; pnictogen included through hoppings; direct (Fe-Fe) + indirect (Fe-pnictogen-Fe) hoppings

✓ Symmetry of the orbitals considered through Slater-Koster parameters to describe the hoppings (pdσ, pdπ, ddσ_1, ddπ_1,ddδ_1)

✓ Straightforward change of pnictogen position (angle α)

MJ Calderon, B.V, E Bascones PRB'09

Phonon Hamiltonian



$$H_{\rm ph} = \sum_{\mathbf{q}} \omega_{\mathbf{q}} a^{\dagger}_{\mathbf{q}} a_{\mathbf{q}} + \sum_{\mathbf{k},\mathbf{q},\mu,\nu,\sigma,M} g^{M}_{\mu\nu}(\mathbf{k},\mathbf{q}) c^{\dagger}_{\mathbf{k}+\mathbf{q}\mu\sigma} c_{\mathbf{k}\nu\sigma} \left(a_{\mathbf{q}} + a^{\dagger}_{-\mathbf{q}} \right)$$

We are just interested in Raman processes i.e **q**=0

The electron-phonon coupling is split in two contributions g^{α} (geometrical) y g^{pd} (through energy integrals pdσ and pdπ). Both have local and non-local contributions:

Geometrical phonon coupling:

$$g^{\alpha,\mathrm{non-loc}}_{\mu\nu}(\mathbf{k}) = \sum_r F^r_{\mu\nu}(\mathbf{k}) \frac{\partial t^r_{\mu\nu}}{\partial \alpha} \delta\alpha \qquad g^{\alpha,\mathrm{loc}}_{\mu\mu} = \frac{\partial \epsilon^{ind}_{\mu}}{\partial \alpha} \delta\alpha$$

Energy integral phonon coupling:

$$\begin{cases} g^{pd,\mathrm{non-loc}}_{\mu\nu}(\mathbf{k}) = \sum_r F^r_{\mu\nu}(\mathbf{k}) \delta pd\sigma \left(\frac{\partial t^r_{\mu\nu}}{\partial pd\sigma} + \frac{\partial t^r_{\mu\nu}}{\partial pd\pi} \frac{pd\pi_0}{pd\sigma_0} \right) \\ g^{pd,\mathrm{loc}}_{\mu\mu} = \delta pd\sigma \left(\frac{\partial \epsilon^{ind}_{\mu}}{\partial pd\sigma} + \frac{\partial \epsilon^{ind}_{\mu}}{\partial pd\pi} \frac{pd\pi_0}{pd\sigma_0} \right) \end{cases}$$

With: $\delta pd\sigma = pd\sigma_0 \frac{1}{f(R_0)} \frac{\partial f(R)}{\partial R} \frac{\partial R}{\partial h} \delta h$

We assume: $f(R) = 1/R^4$ valid for p & d orbitals localized

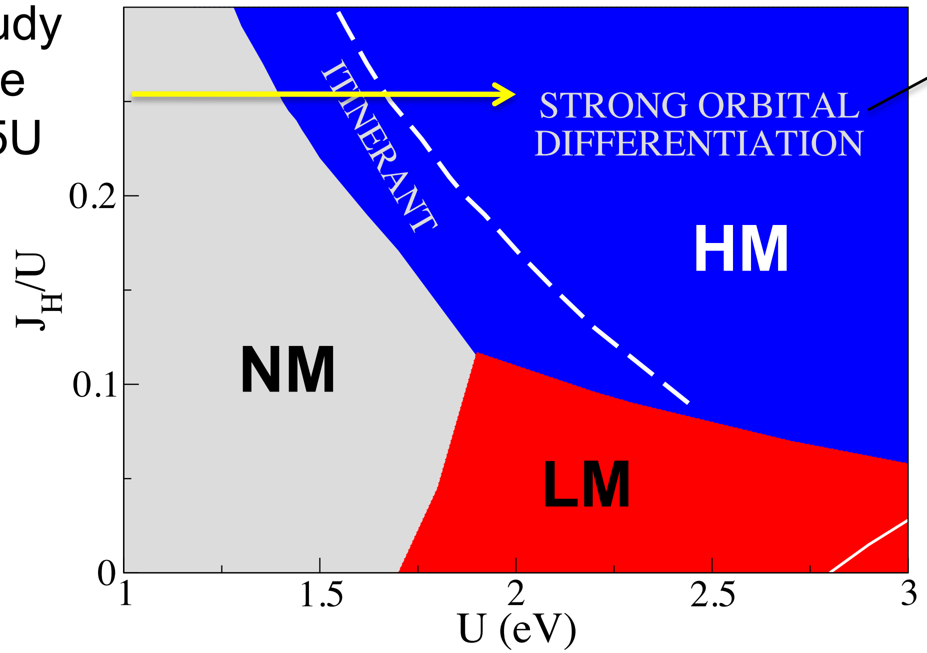
BNL, New York 2013

(π,0) Magnetic phase diagram

$$H = H_{TB} + H_{\text{int}}(U, J)$$

We calculate the **magnetic U-J_H/U phase diagram** applying mean field theory to the *electronic* Hamiltonian.

We study this line J=0.25U

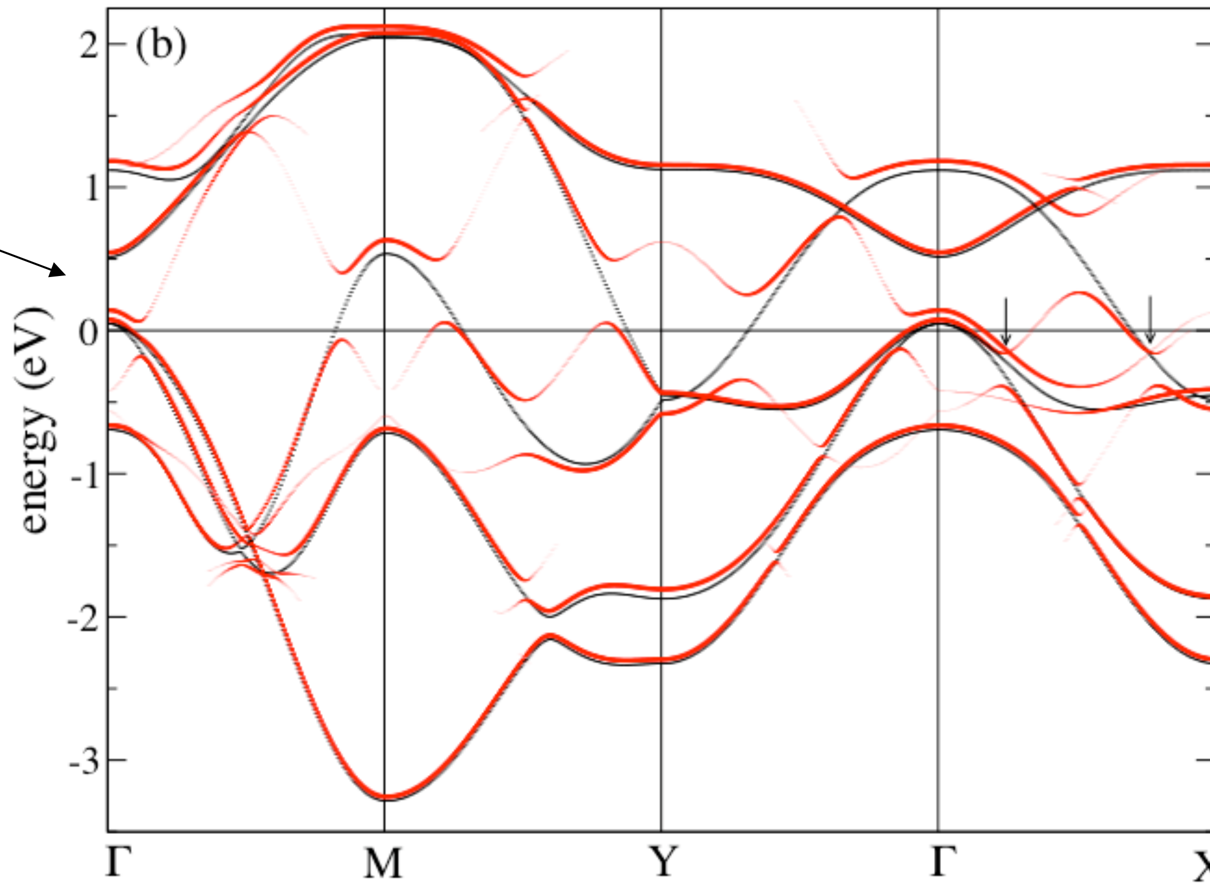


xy and yz become **half-filled gap states**

E. Bascones, M.J. Calderón, B. V., PRL'10; PRB'12

Band structure for the non-magnetic and magnetic regime in the itinerant region

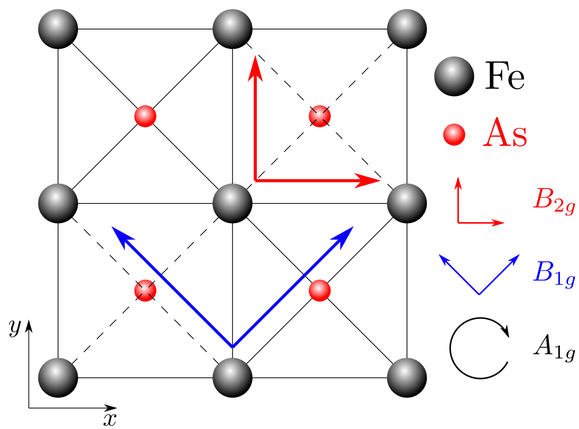
Spectral weight reorganization at low energies due to magnetism in a multiorbital system



We have add a renormalization factor of 3 (in accordance with ARPES)

Raman response for the A1g As-phonon

Raman vertices:



$$\gamma_{\mu\nu}^{B_{1g}}(\mathbf{k}) = \frac{\partial^2 \epsilon_{\mu\nu}(\mathbf{k})}{\partial k_x^2} - \frac{\partial^2 \epsilon_{\mu\nu}(\mathbf{k})}{\partial k_y^2}, \text{ (antisymmetric } k_x \rightarrow k_y\text{)}$$

$$\gamma_{\mu\nu}^{B_{2g}}(\mathbf{k}) = \frac{\partial^2 \epsilon_{\mu\nu}(\mathbf{k})}{\partial k_x \partial k_y}, \text{ (antisymmetric } k_x \rightarrow -k_x \text{ or } k_y \rightarrow -k_y\text{)}$$

$$\gamma_{\mu\nu}^{A_{1g}}(\mathbf{k}) = \frac{\partial^2 \epsilon_{\mu\nu}(\mathbf{k})}{\partial k_x^2} + \frac{\partial^2 \epsilon_{\mu\nu}(\mathbf{k})}{\partial k_y^2}.$$

In the PM state B1g and B2g Raman response are zero because they are orthogonal to the symmetry of the A1g As-phonon.

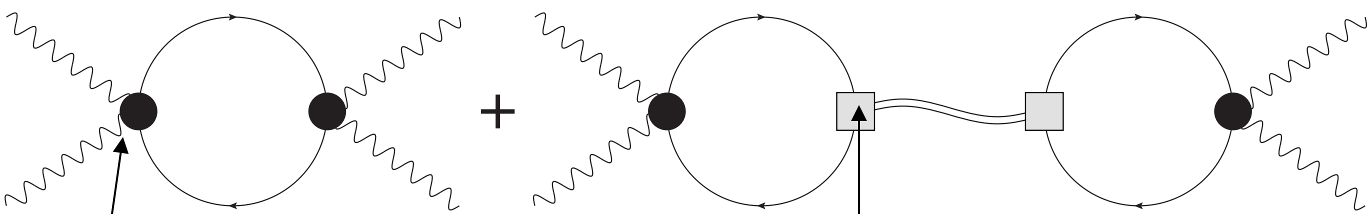
Our proposal: Due to the electron-phonon coupling the B1g Raman response of the A1g As-phonon is large in the magnetic state since the tetragonal symmetry is broken by the $(\pi,0)$ magnetic ordering:

Anomalies in phonons driven by magnetism

Charge-phonon theory I:

The Raman response is calculated using the Charge-phonon theory

$$\chi^{\lambda}(\Omega) = \chi^{\lambda}_{el-el}(\Omega) + \Delta\chi^{\lambda}_{ph}(\Omega)$$



Raman vertex

Electron phonon Interaction

$$\chi^{\lambda}_{el-el}(\Omega) \qquad \Delta\chi^{\lambda}_{ph,M,M'}(\Omega) = \chi^{\lambda}_{M}(\Omega) D_0(\Omega) \chi^{*\lambda}_{M'}(\Omega)$$

This contribution has been calculated in B.V, Calderon, Leon, Bascones PRB'13

Our main calculation

Mixed bubble

Phonon propagator

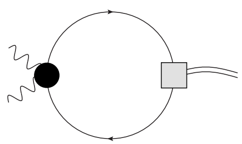
$$D_0(\Omega) = \frac{1}{(\Omega - \Omega_0) + i\Gamma_0}$$

Ω_0 and Γ_0 are the phonon frequency and the phonon scattering rate

Charge-phonon theory II:

$$\Delta\chi^{\lambda}_{ph,M,M'}(\Omega) = \chi^{\lambda}_{M}(\Omega) D_0(\Omega) \chi^{*\lambda}_{M'}(\Omega)$$

$$\chi^{\lambda}_{M}(\Omega)$$

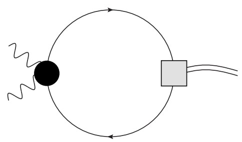


$$\chi''^{\lambda}_{M}(\Omega) = -\frac{\pi}{V} \sum_{\mathbf{k}\sigma nn'} \gamma^{\lambda}_{nn'}(\mathbf{k}) g^{M*}_{nn'}(\mathbf{k}) \left(f(E_n(\mathbf{k})) - f(E_{n'}(\mathbf{k})) \right)$$
$$\times \left(\delta(\Omega + E_n(\mathbf{k}) - E_{n'}(\mathbf{k})) - \delta(-\Omega + E_n(\mathbf{k}) - E_{n'}(\mathbf{k})) \right)$$

Charge-phonon theory II:

$$\Delta\chi^{\lambda}_{ph,M,M'}(\Omega) = \chi^{\lambda}_{M}(\Omega) D_0(\Omega) \chi^{*\lambda}_{M'}(\Omega)$$

$$\chi^{\lambda}_{M}(\Omega)$$



$$\chi''^{\lambda}_{M}(\Omega) = -\frac{\pi}{V} \sum_{\mathbf{k}\sigma nn'} \gamma^{\lambda}_{nn'}(\mathbf{k}) g^{M*}_{nn'}(\mathbf{k}) \left(f(E_n(\mathbf{k})) - f(E_{n'}(\mathbf{k}))\right) \times \left(\delta(\Omega + E_n(\mathbf{k}) - E_{n'}(\mathbf{k})) - \delta(-\Omega + E_n(\mathbf{k}) - E_{n'}(\mathbf{k}))\right)$$

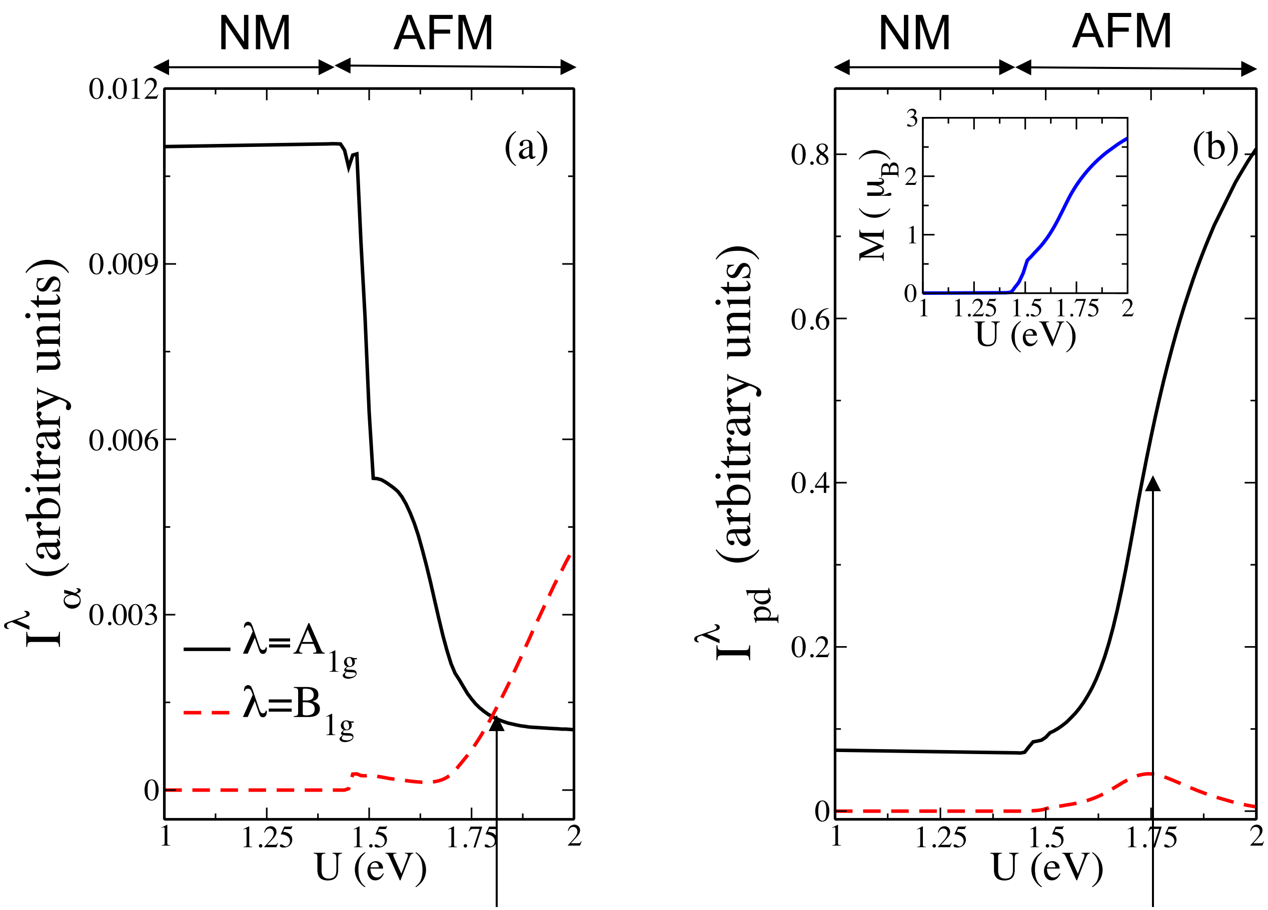
$$\mathrm{Im}\Delta\chi^{\lambda}_{ph,M}(\Omega) = -I^{\lambda}_{M} \frac{(q^{\lambda}_{M})^2 - 1 + 2\left(\frac{\Omega-\Omega_0}{\Gamma_0}\right) q^{\lambda}_{M}}{(q^{\lambda}_{M})^2 \left(1 + \left(\frac{\Omega-\Omega_0}{\Gamma_0}\right)^2\right)}$$

$$I^{\lambda}_{M} = \frac{\left(\chi'^{\lambda}_{M}(\Omega_0)\right)^2}{\Gamma_0} \qquad q^{\lambda}_{M} = -\frac{\chi'^{\lambda}_{M}(\Omega_0)}{\chi''^{\lambda}_{M}(\Omega_0)}$$

Charge phonon theory related to Fano theory. All the physics is encoded in the mixed bubble:

E. Cappelluti, et al, PRB'12

Results: A1g As-phonon Raman intensity



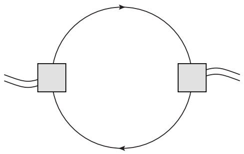
But the resulting intensity cannot be calculated due to uncertainties in the values of the phonon couplings g_{α} and g_{pd}

There is a critical value of U where $I_{B1g} > I_{A1g}$ as in $BaFe_2As_2$

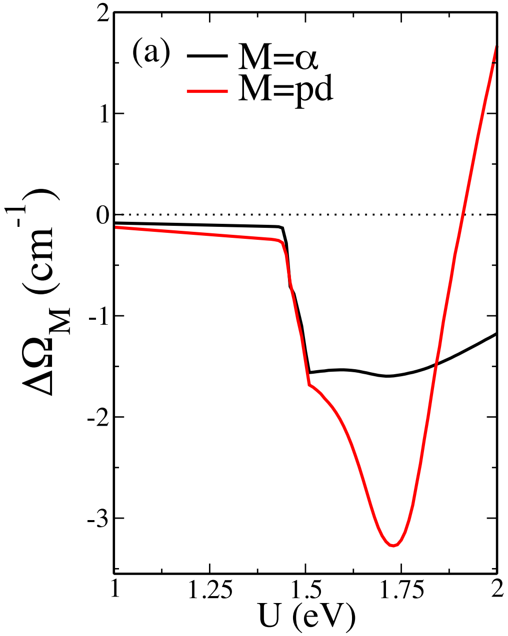
I_{A1g} increases a lot in the magnetic state as in 122 compounds

BNL, New York 2013

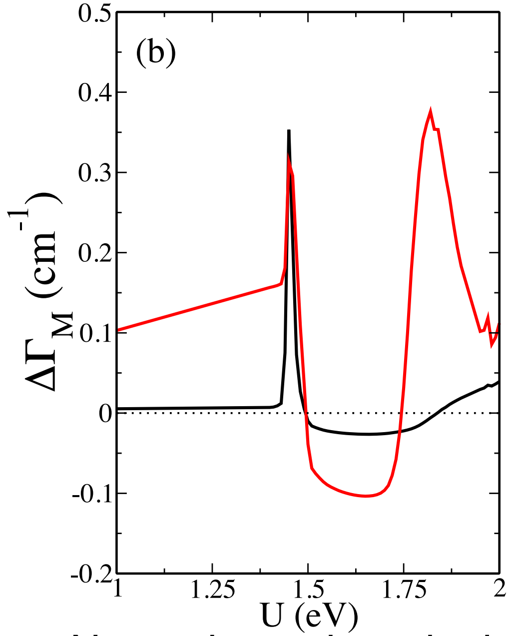
Phonon hardening/softening and life-time



$$\Delta\Omega_M = \Pi'_M(\Omega_0, U) - \Pi'_M(\Omega_0, U=0) \quad \Delta\Gamma_M = -(\Pi''_M(\Omega_0, U) - \Pi''_M(\Omega_0, U=0)$$



Hardening just for large values of U when e-ph coupling goes via g_{pd}

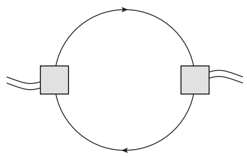


Softening of the phonon frequency in the magnetic state.

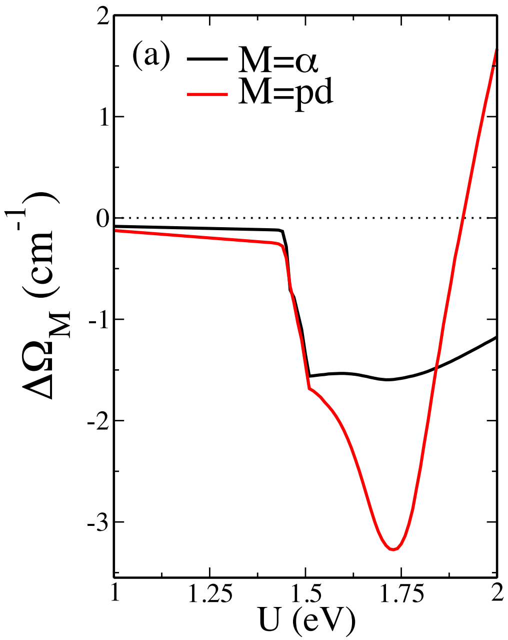
Narrowing or broadening appear depending on parameters

BNL, New York 2013

Phonon hardening/softening and life-time



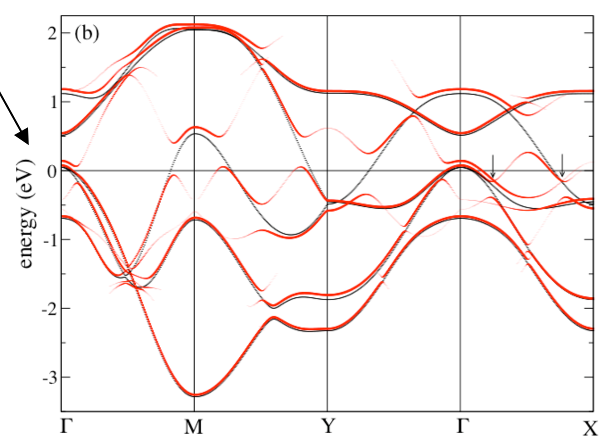
$$\Delta\Omega_M = \Pi'_M(\Omega_0, U) - \Pi'_M(\Omega_0, U=0) \qquad \Delta\Gamma_M = -(\Pi''_M(\Omega_0, U) - \Pi''_M(\Omega_0, U=0)$$

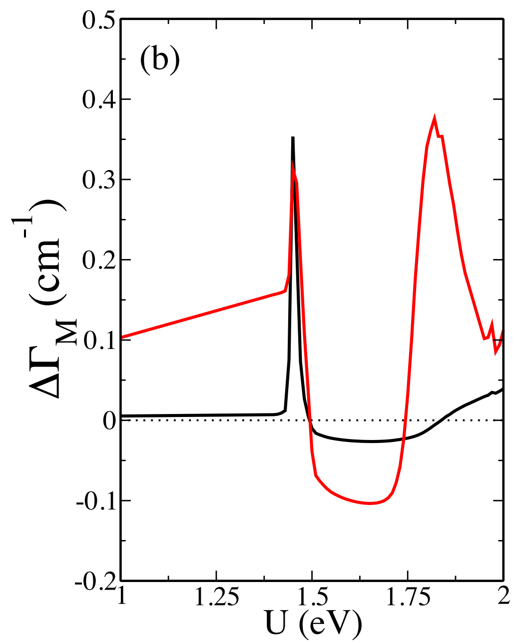


Softening of the phonon frequency in the magnetic state.

Hardening just for large values of U when e-ph coupling goes via g_{pd}

The softening is related to the spectral weight redistribution in a multiorbital system from higher energies ($\Omega > \Omega_0$) to lower energies ($\Omega < \Omega_0$) when entering into the magnetic state.





Narrowing or broadening appear depending on parameters

BNL, New York 2013

Summary of our work

- The electron-phonon coupling is via the dependence of the Slater-Koster parameters (α, pdσ, pdπ) on the As position.
- Magnetism is included at the mean-field Hartree-Fock level.
- The Raman response is evaluated in the paramagnetic and in the (π, 0) magnetic states using the proper generalization of the charge-phonon theory.

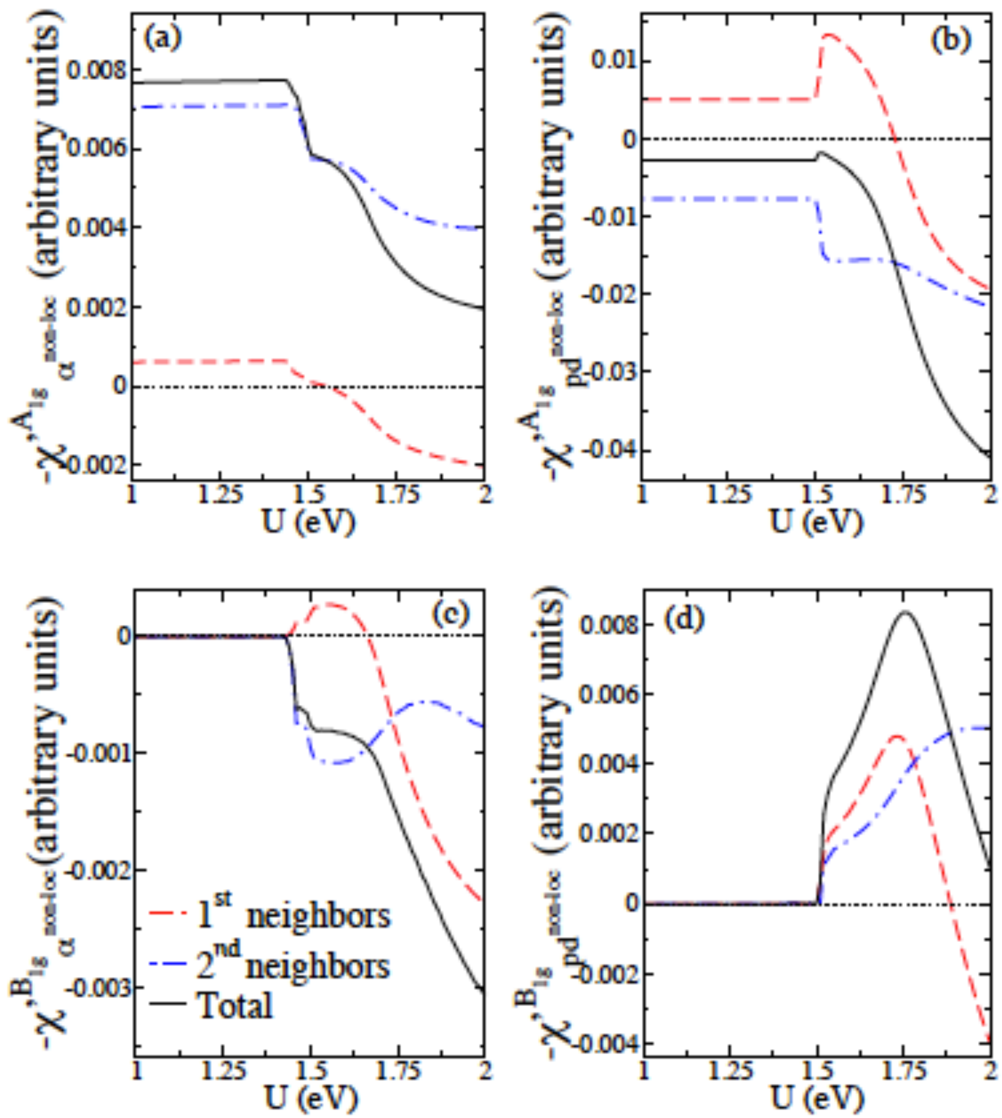
- A finite Raman intensity can be observed in the magnetic state in the B1g but not in the B2g polarization and it is a consequence of the coupling of the phonons to the anisotropic (π,0) magnetic state.
- Softening and hardening are possible.
- For a quantitative comparison **more work is needed.**

Outlook

- **It is possible that in the nematic state there is a signal in B_{1g}.**
- **In the double stripe magnetic state of FeTe, the out-of-plane Te-phonon will be different from zero in the B2g polarization geometry, instead B1g symmetry.**

It would be interesting to explore these possibilities experimentally.

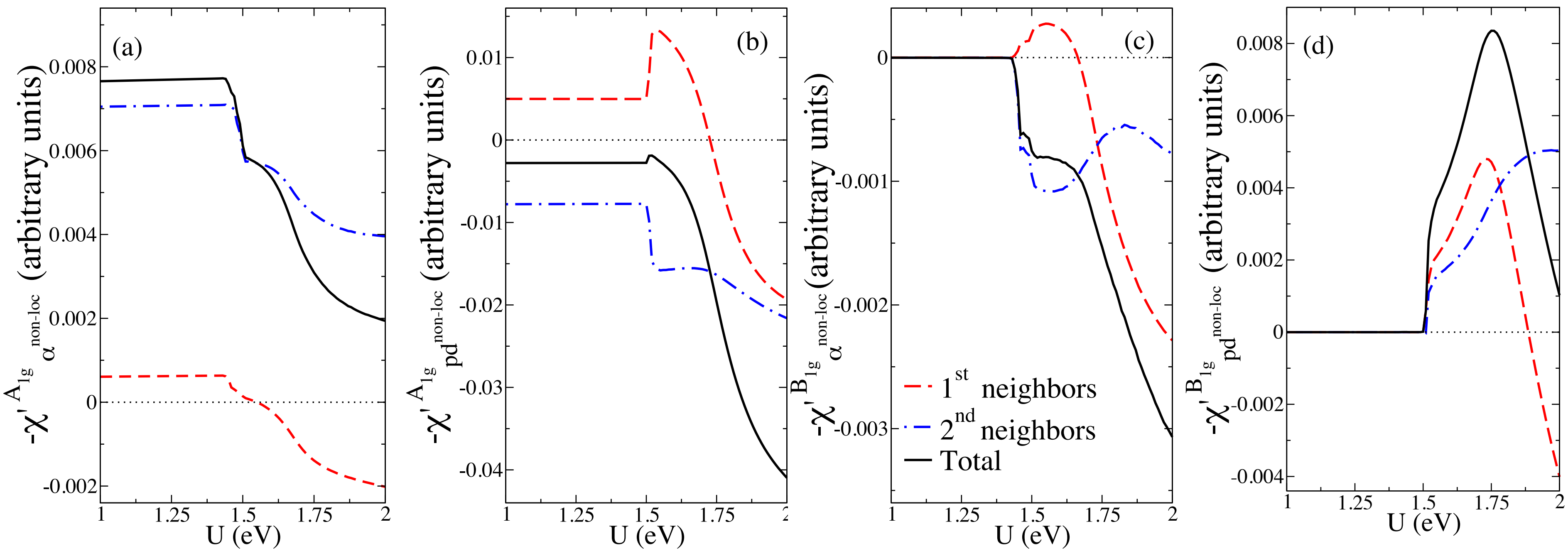
Thank you!



(a)
(b)
(c)
(d)
U (eV)
U (eV)
U (eV)
U (eV)
$-\chi'^{A_{1g}}_{\alpha}{}^{non\text{-}loc}$ (arbitrary units)
$-\chi'^{A_{1g}}_{pd}{}^{non\text{-}loc}$ (arbitrary units)
$-\chi'^{B_{1g}}_{\alpha}{}^{non\text{-}loc}$ (arbitrary units)
$-\chi'^{B_{1g}}_{pd}{}^{non\text{-}loc}$ (arbitrary units)
1st neighbors
2nd neighbors
Total

Going beyond....

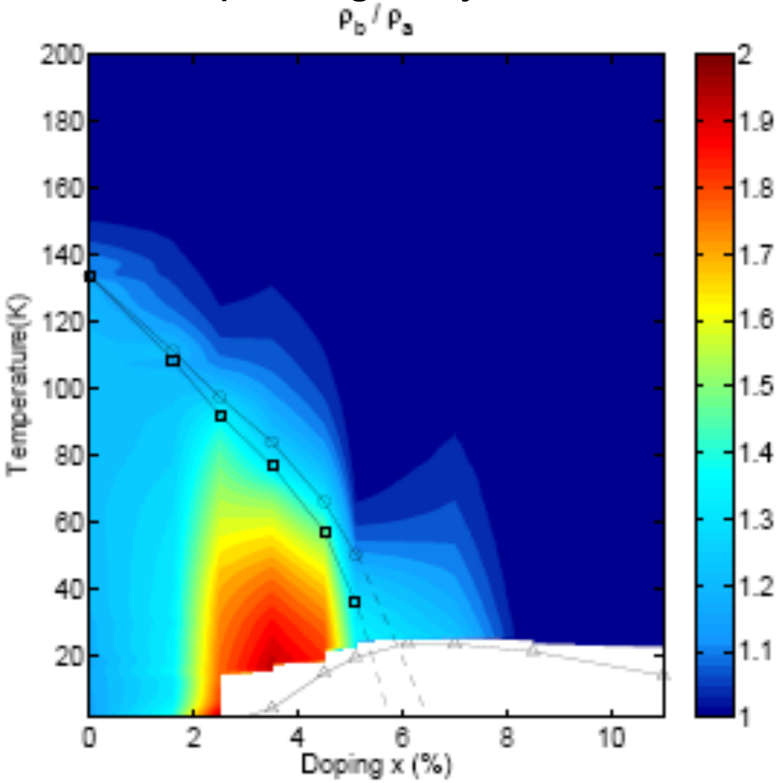
- The interplay between magnetism and As-heigh should be treated self-consistently
- The electron-phonon coupling could be go through the interactions
- Magnetism beyound mean-field.



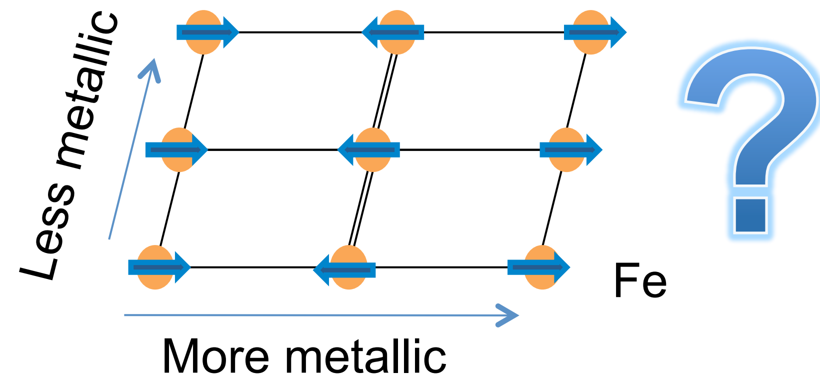
(a)
(b)
(c)
(d)
U (eV)
$-\chi'^{A_{1g}}_{\alpha}{}^{non-loc}$ (arbitrary units)
$-\chi'^{A_{1g}}_{pd}{}^{non-loc}$ (arbitrary units)
$-\chi'^{B_{1g}}_{\alpha}{}^{non-loc}$ (arbitrary units)
$-\chi'^{B_{1g}}_{pd}{}^{non-loc}$ (arbitrary units)
1st neighbors
2nd neighbors
Total

Anisotropy in transport:

-Anisotropy in transport (Chu et al. PRB'10; Science'10, Tanatar et al. PRB'11) and in optical conductivity (Dusza et al, EPL'11, Review: I.R. Fisher et al. Rep. Prog. Phys.'11, Nakakima et al. '11), etc



Chu et al. Science'10

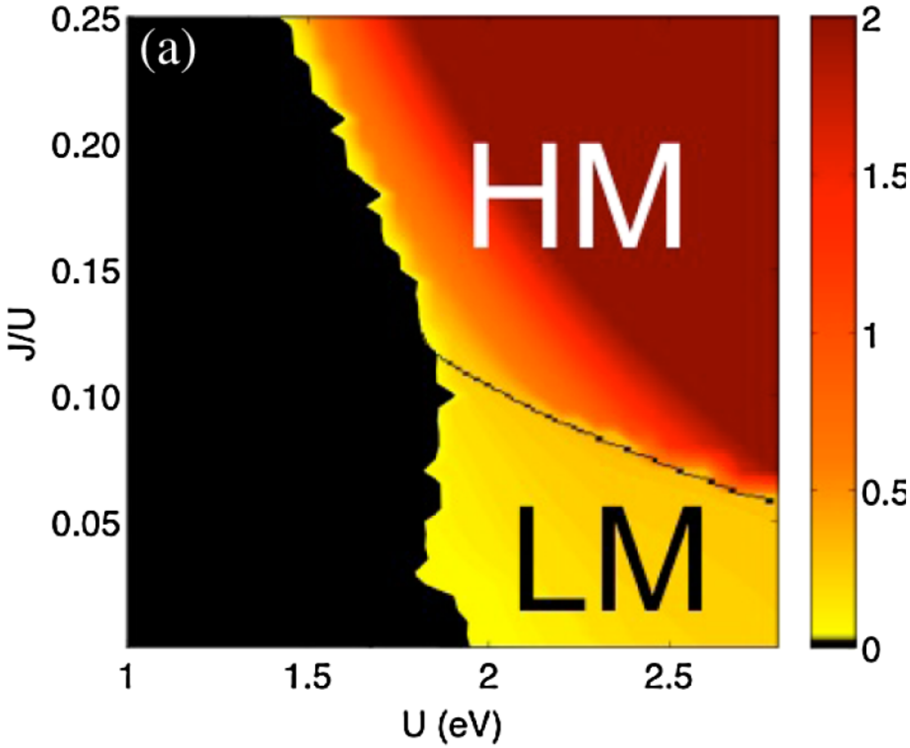


Unexpected because:

- FM bond is shorter than AF one
- AF is naively associated with gaps and FM with metallicity
- Scattering rate is larger in the AF direction

BNL, New York 2013

Magnetism: (π,0) mean field phase diagram: metallic region



Drude weight ratio

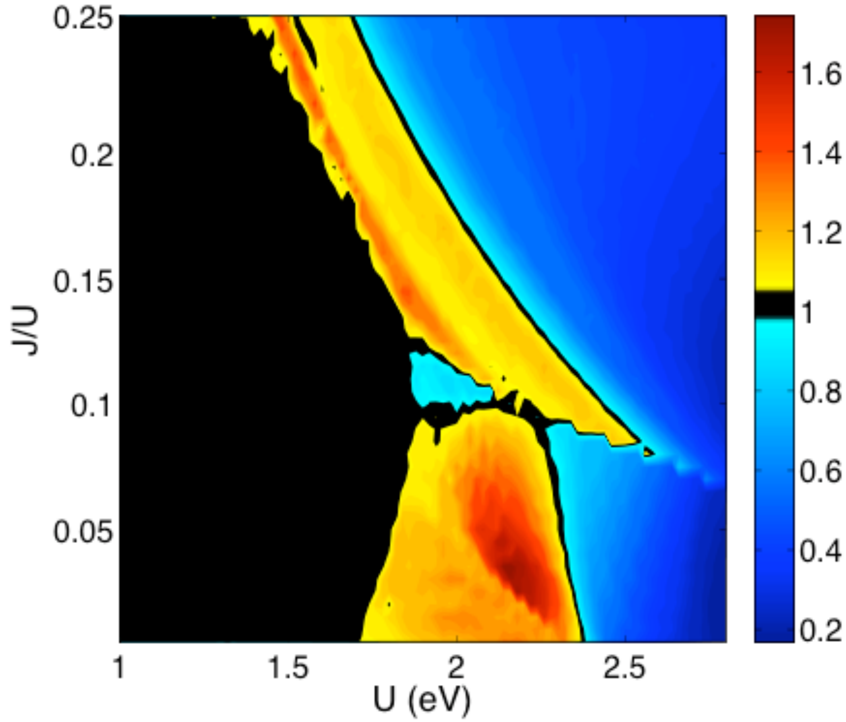
$$D_a / D_b = \frac{\sum_{k,n} v_a^2(\mathbf{k},n)\, g(\mathbf{k},n)\, \delta(\varepsilon_n(\mathbf{k}) - E_F)}{\sum_{k,n} v_b^2(\mathbf{k},n)\, g(\mathbf{k},n)\, \delta(\varepsilon_n(\mathbf{k}) - E_F)}$$

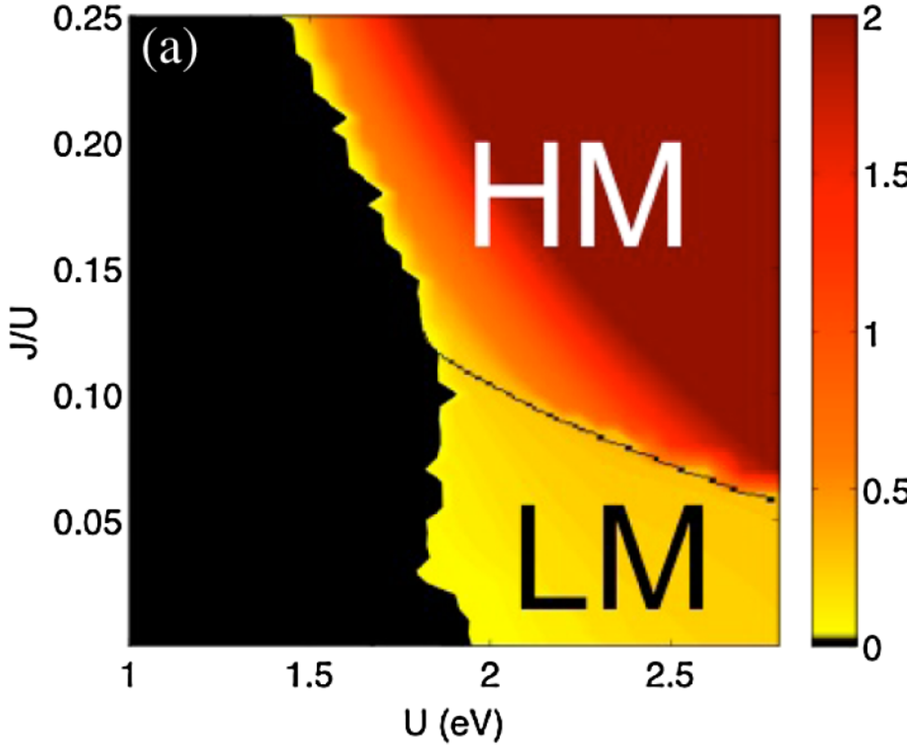
We also calculate the Drude ratio with the Kubo formula and get the same result.

We assume the scattering rate is isotropic

Drude weight anisotropy:

Drude weight ratio: D_a/D_b

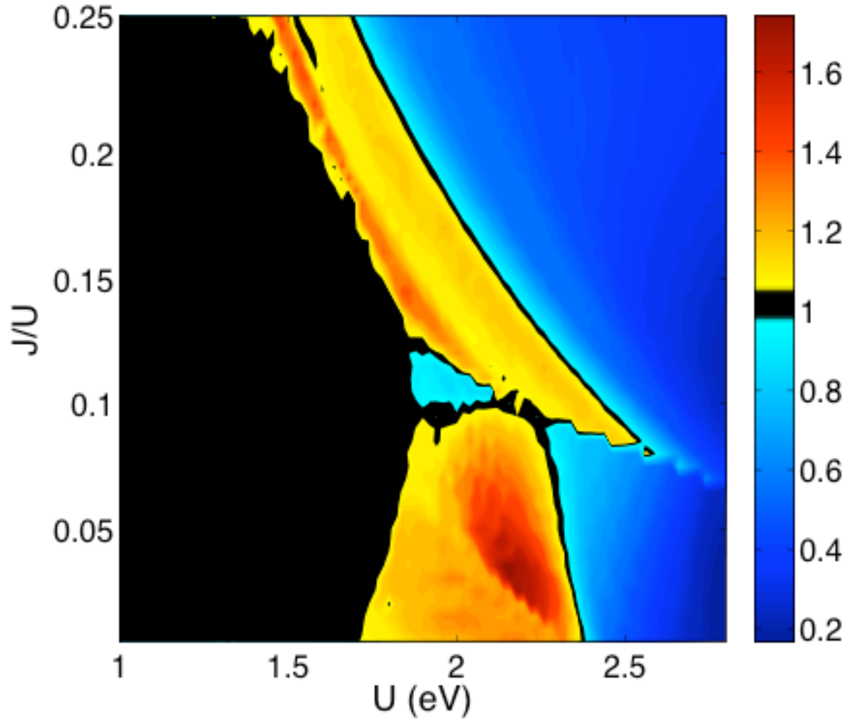




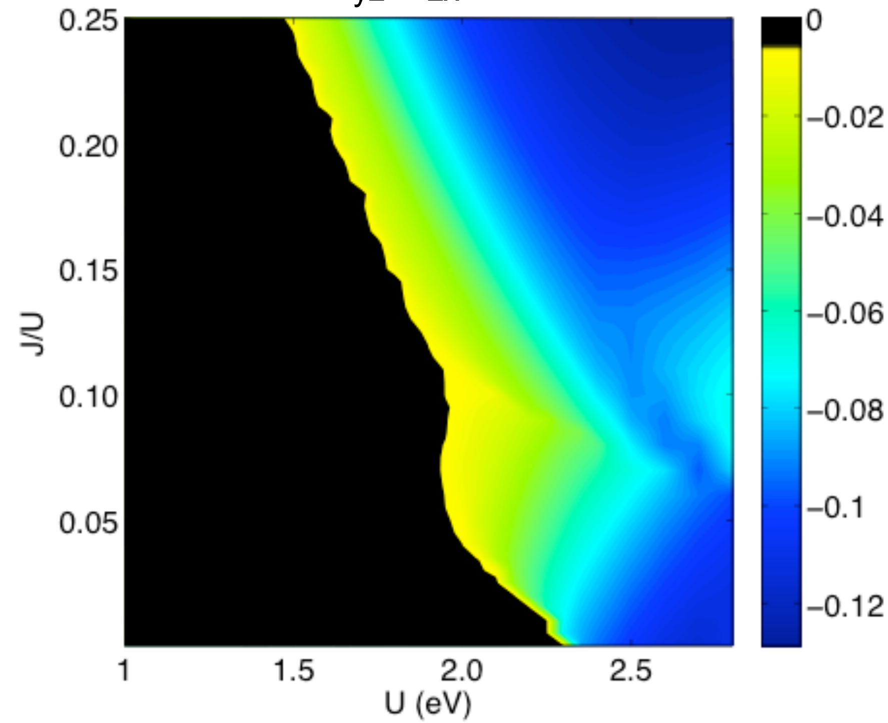
$D_a/D_b>1$ consistent with experimental $r_a/r_b<1$
Is given in regions with low magnetic moment

Is orbital order responsible of resistivity anisotropy?

Drude weight ratio: D_a/D_b



n_{yz}-n_{zx}

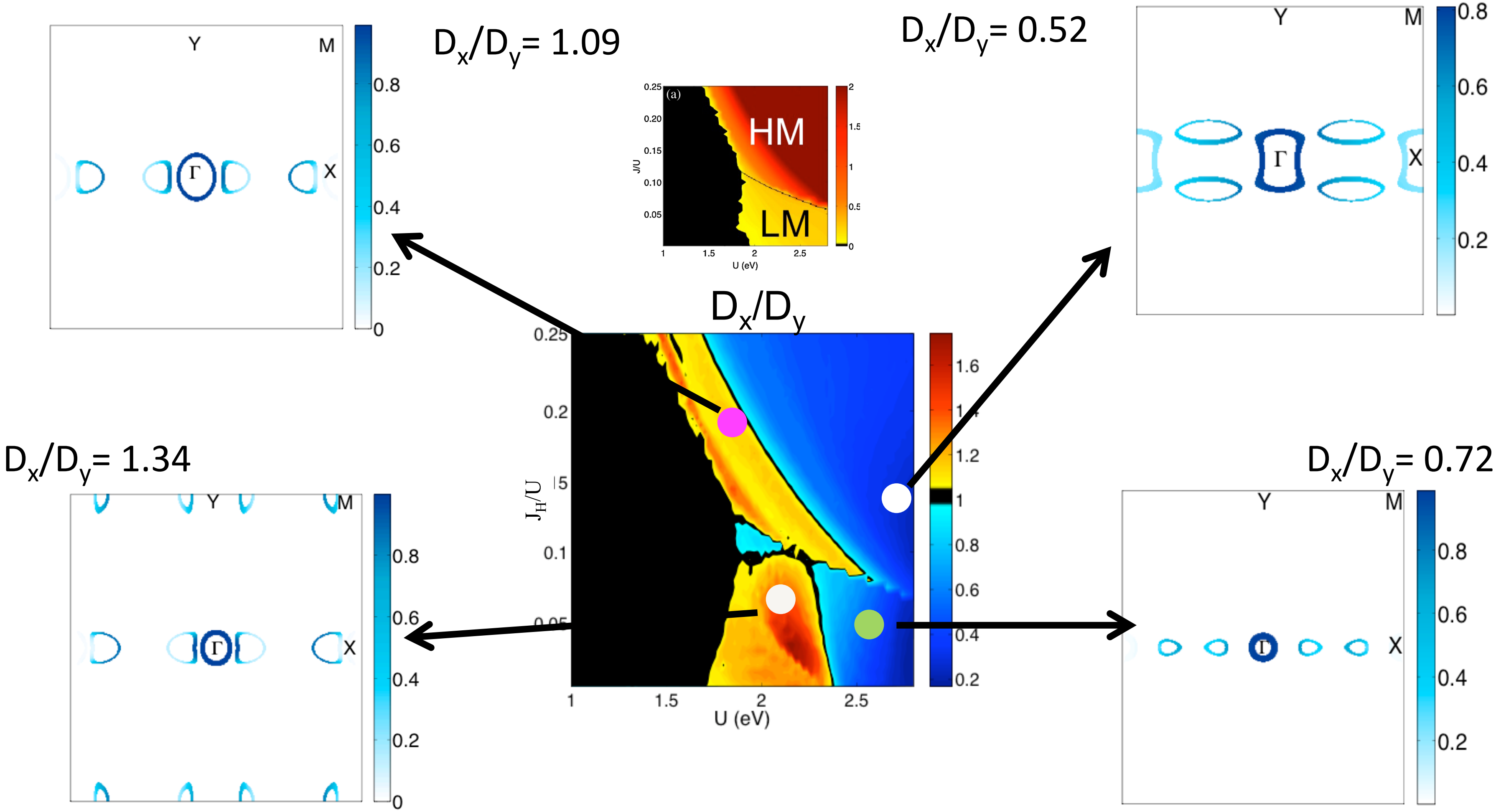


$D_a/D_b>1$ consistent with experimental $r_a/r_b<1$

$n_{yz}<n_{zx}$

Experimental signs of anisotropy and orbital ordering are anticorrelated, B. Valenzuela, E. Bascones, M.J. Calderón, PRL 105, 207202 (2010)

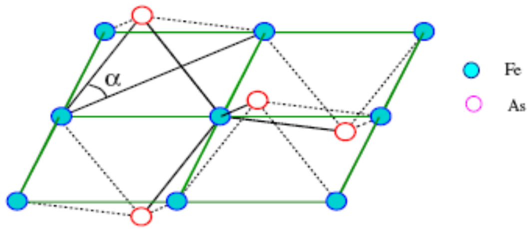
Magnetic reconstruction as origin of the conductivity anisotropy



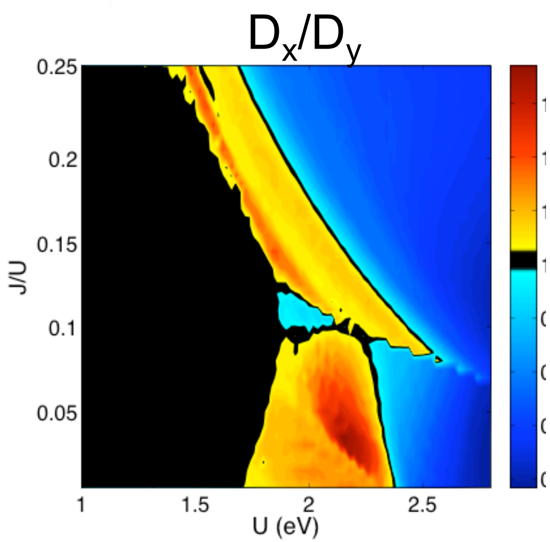
Anisotropy linked to topology and morphology of the Fermi Surface.
Experimental anisotropy in general for low moment.

B. Valenzuela, EB, M.J. Calderón, PRL 105, 207202 (2010)

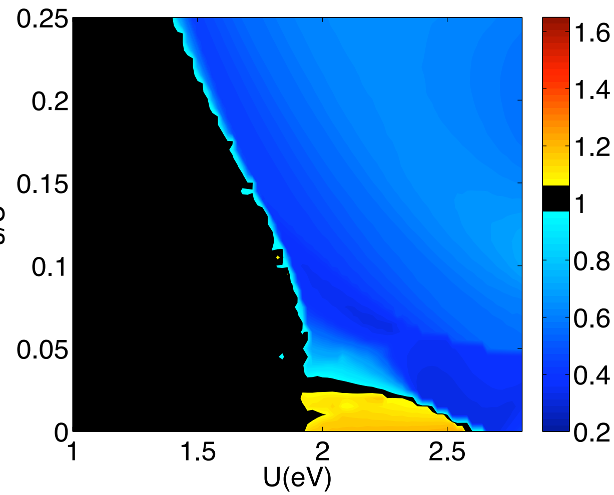
Sensitivity of the anisotropy to the angle α



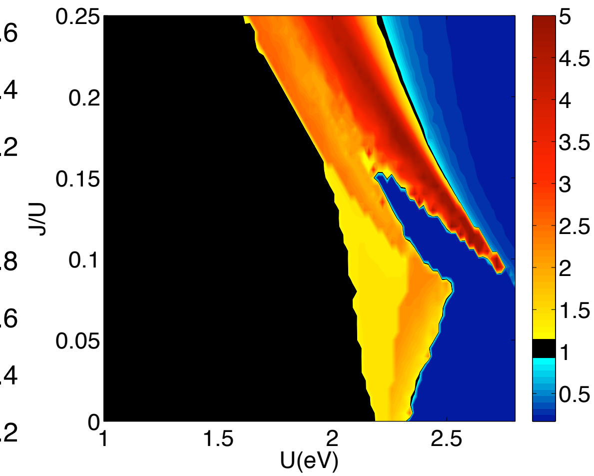
Regular tetrahedron



Squashed tetrahedron



Elongated tetrahedron



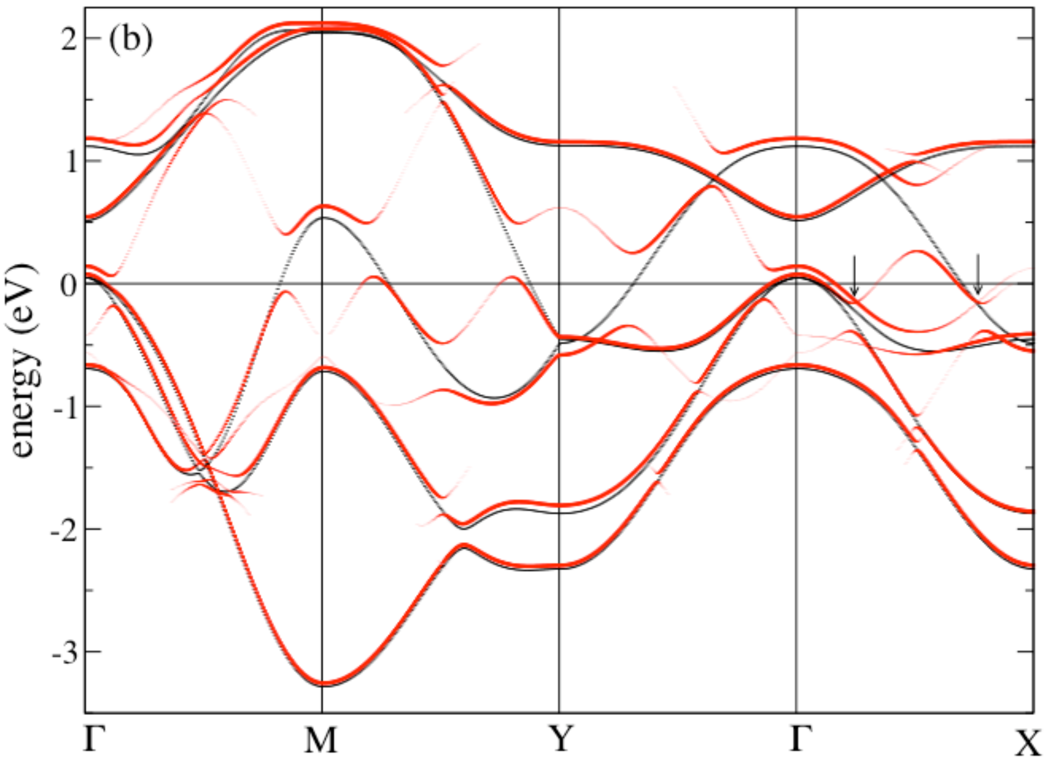
In the squashed tetrahedron the region with Dx/Dy>1 has reduced very much and it is increased for the elongated tetrahedron.

BNL, New York 2013

Bands

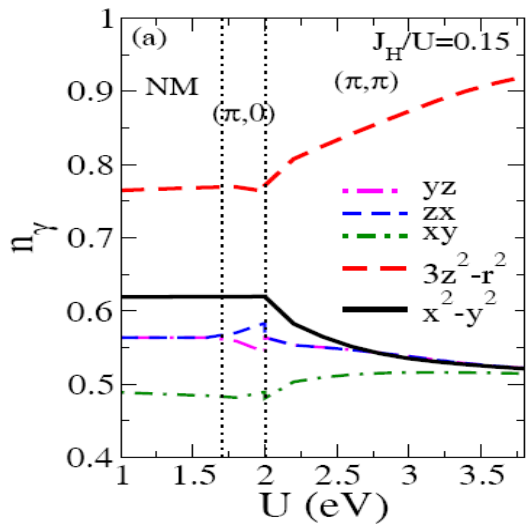
The effect of magnetism is not just at the Fermi surface but it is seen **at all energies in the band**

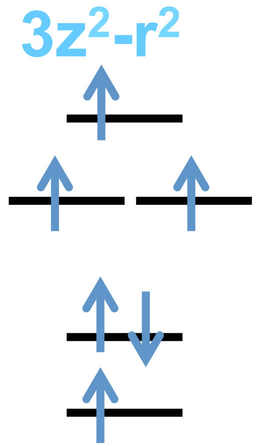
U=2.2, J=0.07U

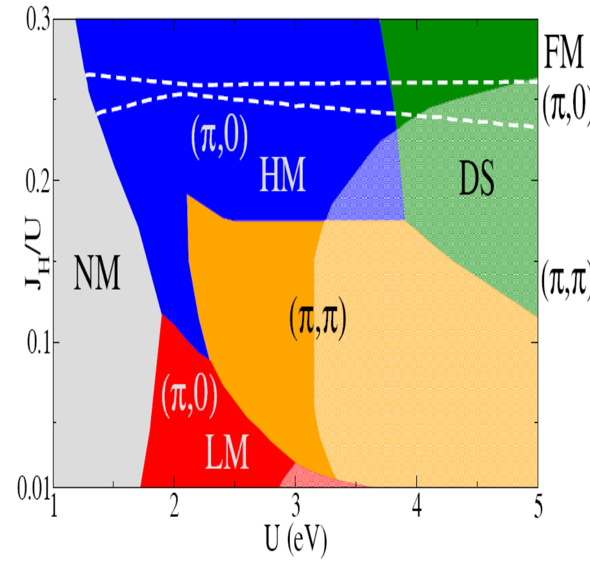


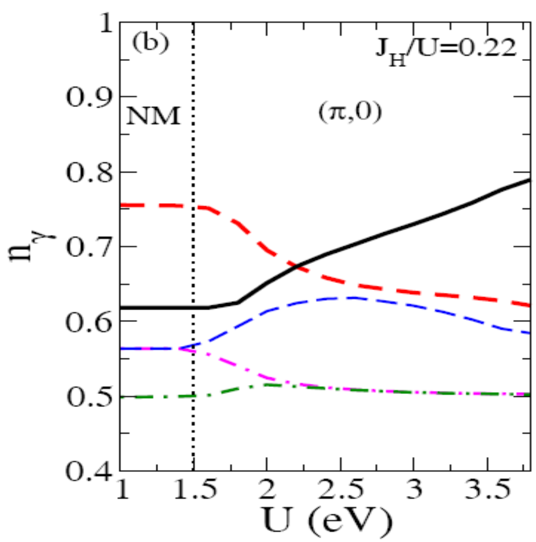
BNL, New York 2013

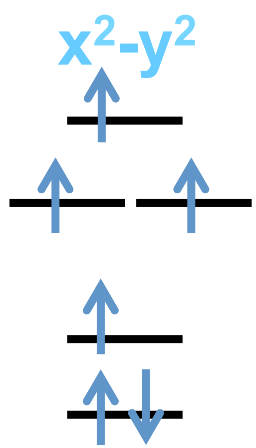
Orbital reorganization in the Hartree-Fock phase diagram









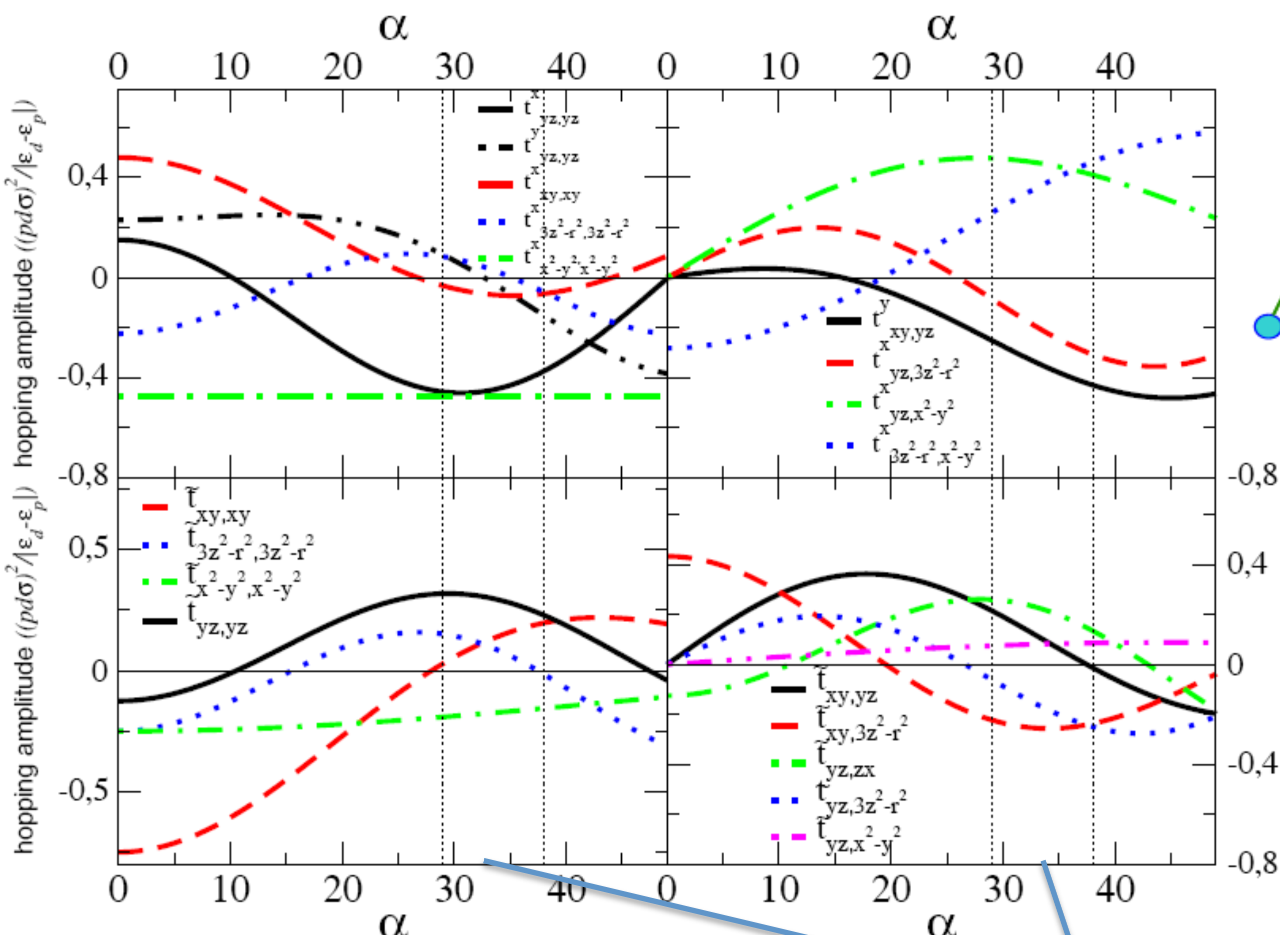


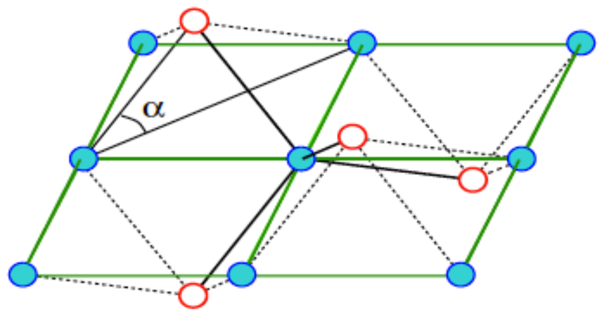
$(\pi,0)$ in x^2-y^2 configuration
(π,π) in $3z^2$-r^2 configuration

Resembles the orbital reorganization found in the strong coupling limit

M.J. Calderón, G. León , B. Valenzuela, EB, arX

Tight-binding for five orbitals: angle dependence of the hoppings





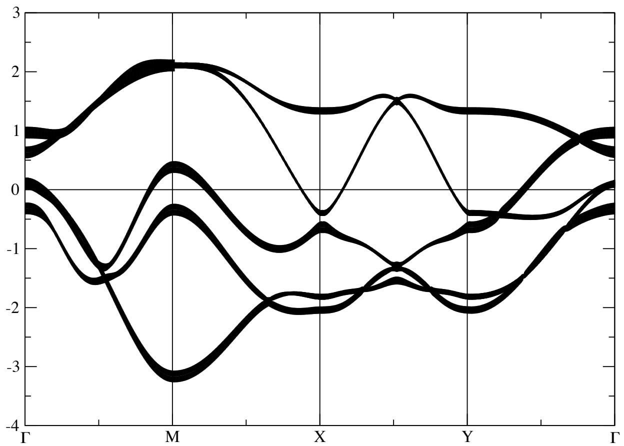
Hoppings related by symmetry and calculated with four fitting parameters

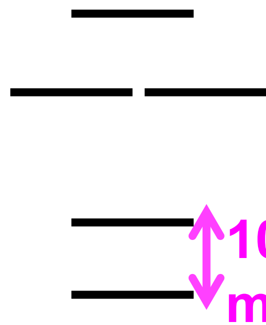
MJ Calderon, B.V, E Bascones PRB'09

Experimental range

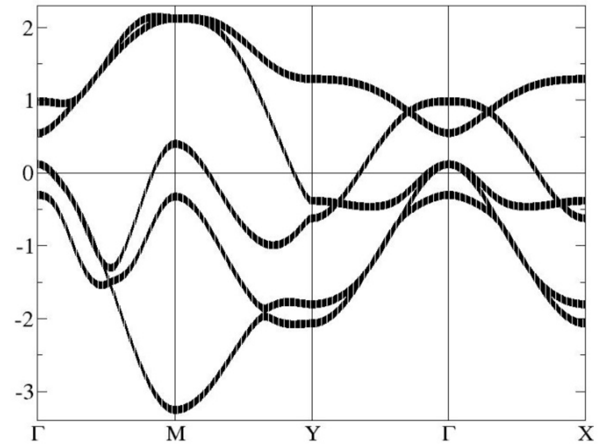
BNL, New York 2013

Hartree-Fock phase diagram. Sentivity to Crystal field

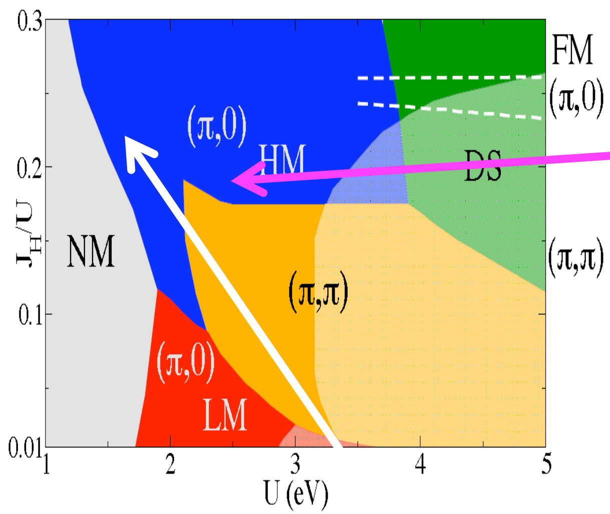




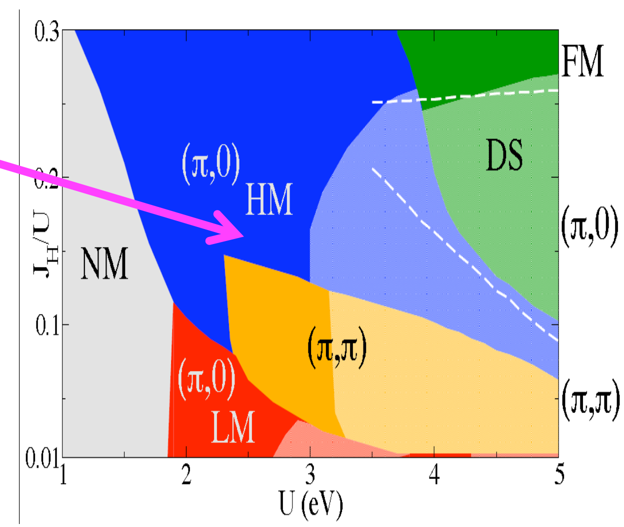
100 [illegible] 50 mev



$3z^2$-r^2 /x^2-y^2 CF 100 meV larger

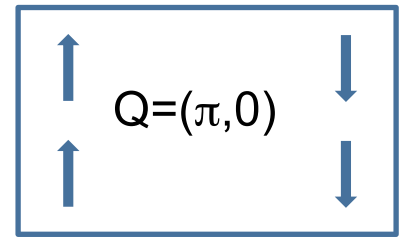


Correlated metal

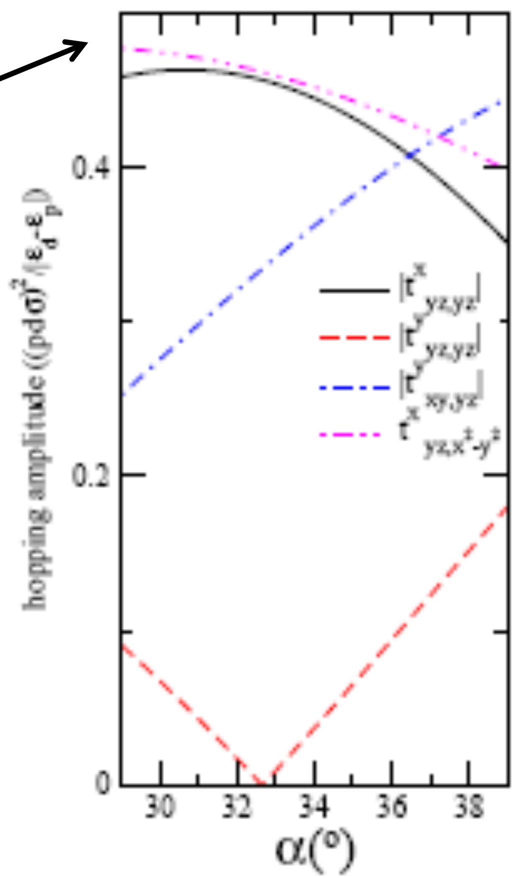


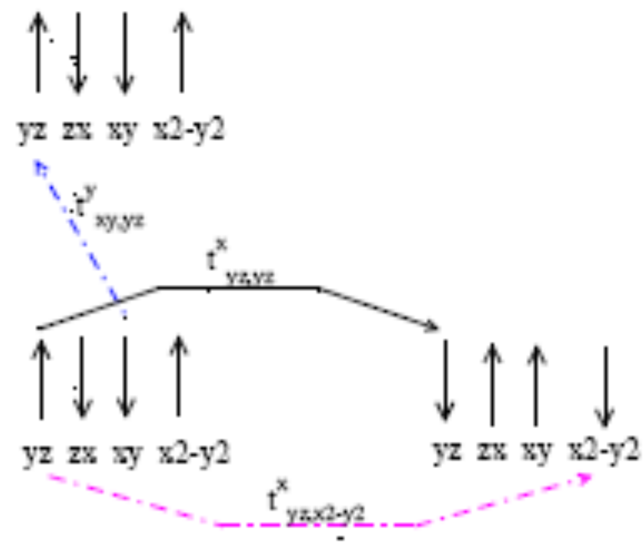
Nesting driven (π,0)

Reasoning from the strong coupling point of view to understand the LM



The low moment phase is stabilized because crossed hoppings are as big as direct hoppings and also very anisotropic: **Release frustration**

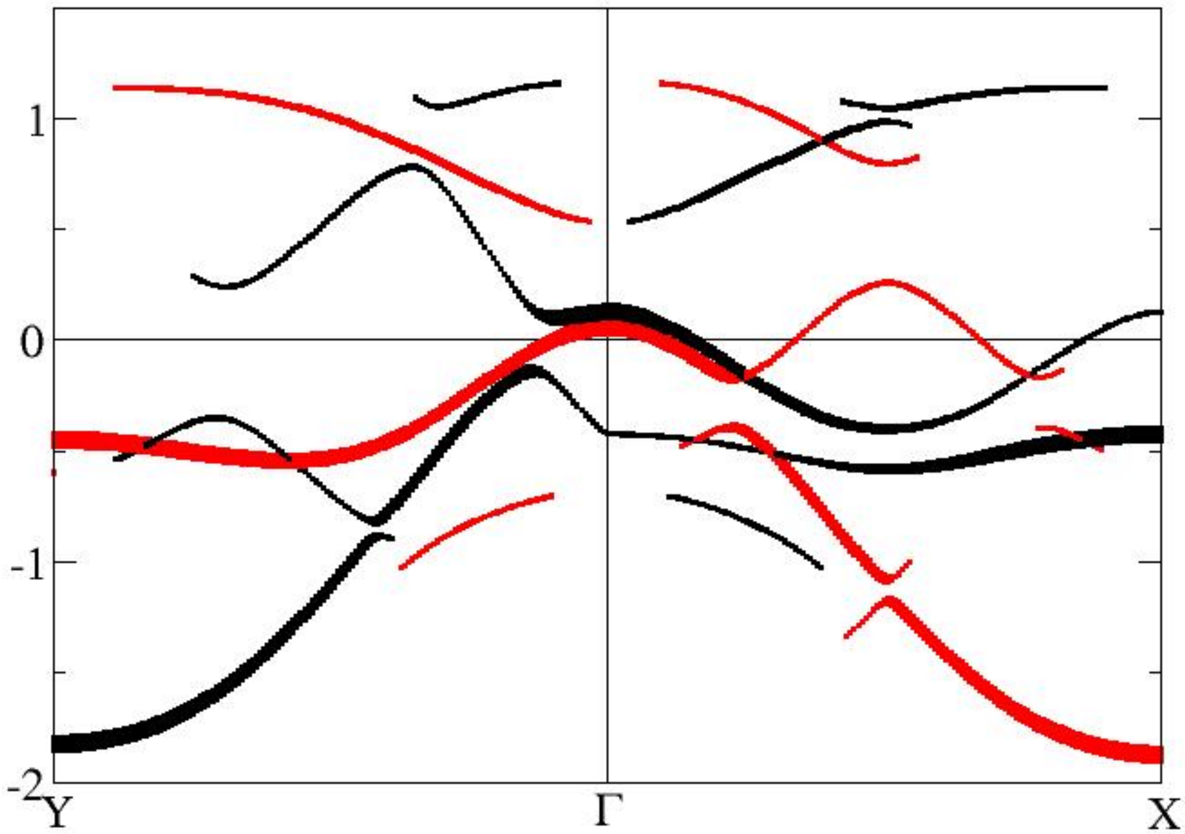




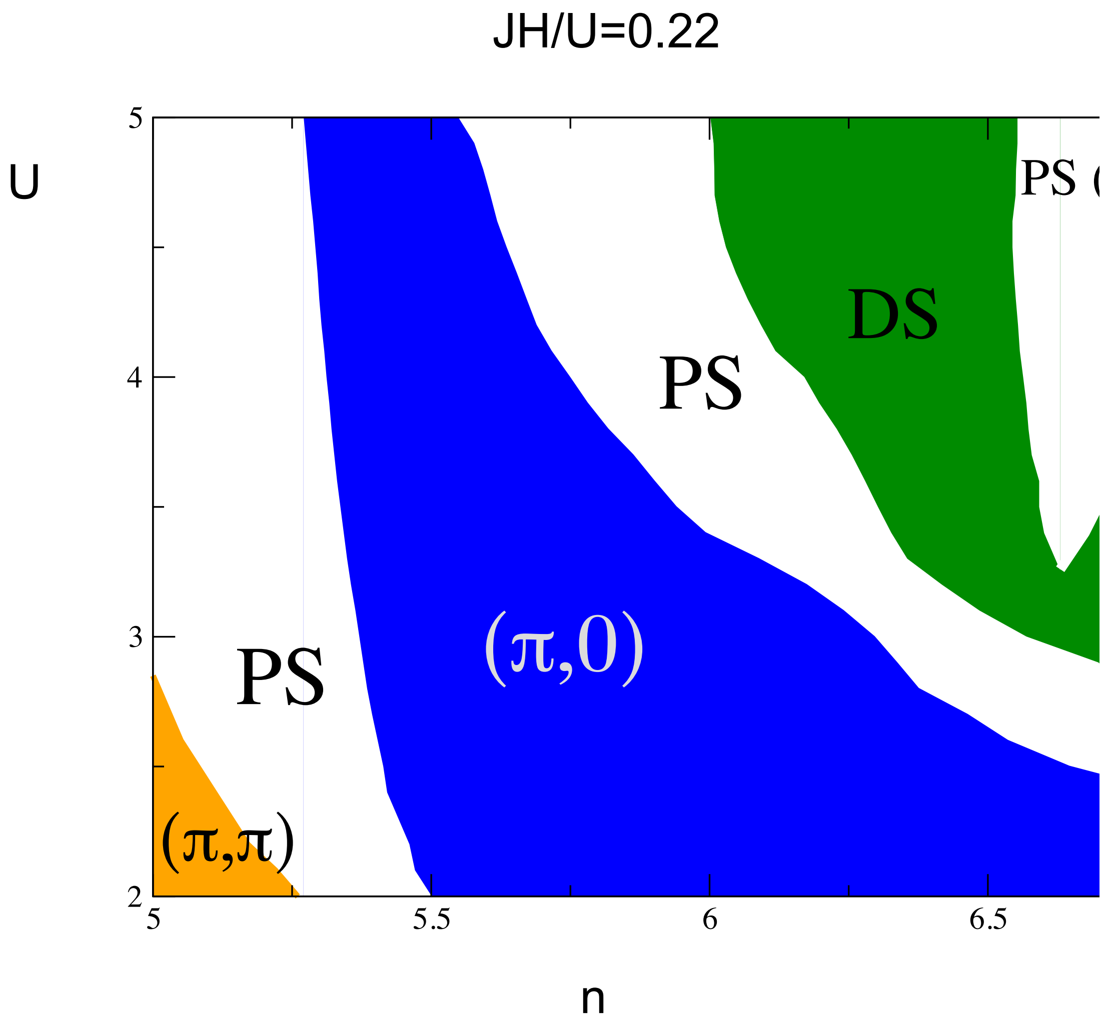
$$t^y_{yz,x^2-y^2} = 0$$

$$t^x_{xy,yz} = 0$$

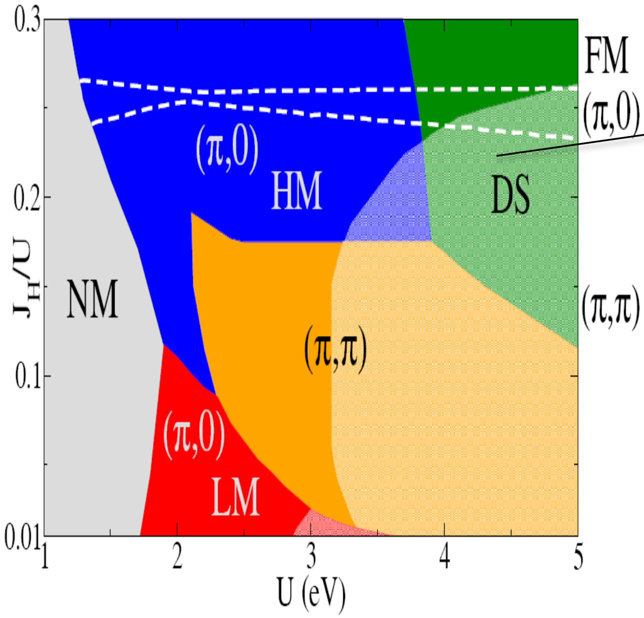
Orbital ordering in the band structure for U=2.2 eV and J=0.07U

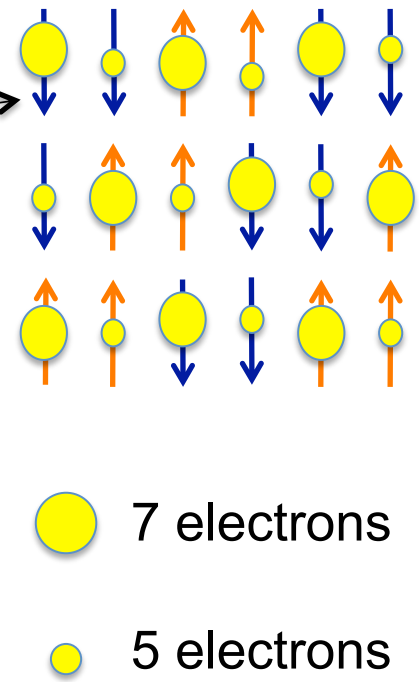


Phase separation

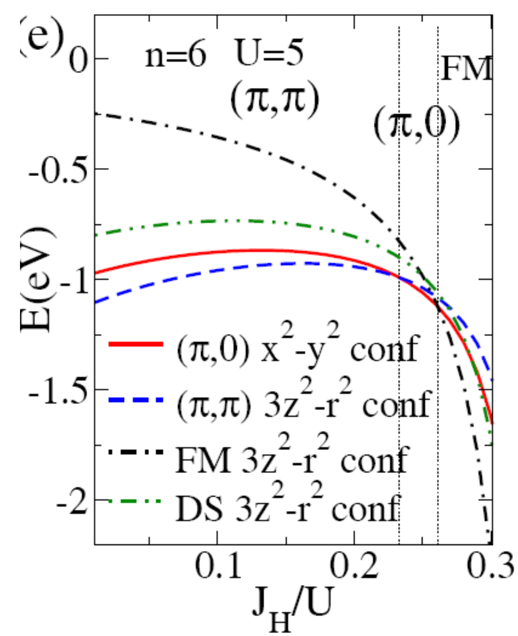


Magnetism: Mean field phase diagram





In the mean field description there is a DS phase charge modulated instead of FM. But strong coupling analysis also points to DS instability at high J_H



M.J. Calderón, G. León , B. V., E. Bascones, arXiv: 1107.2279 (2011)